

# Berry-like phases in structured atoms and molecules

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## Abstract

Quantum mechanical phases arising from a periodically varying Hamiltonian are considered. These phases are derived from the eigenvalues of a stationary, “dressed” Hamiltonian that is able to treat internal atomic or molecular structure in addition to the time variation. In the limit of an adiabatic time variation, the usual Berry phase is recovered. For more rapid variation, non-adiabatic corrections to the Berry phase are recovered in perturbation theory, and their explicit dependence on internal structure emerges. Simple demonstrations of this formalism are given, to particles containing interacting spins, and to molecules in electric fields.

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## I. INTRODUCTION

Any quantum mechanical system in a stationary state accumulates a dynamical phase over time proportional to the energy of that state. To determine energy differences, based on phase differences accumulated between two such states, is the basis of Ramsey spectroscopy; the workhorse of high-precision measurement. For this reason, small effects that can add spurious phase shifts must be understood and kept under control.

One such variety of spurious phases arises when the states of interest are not strictly stationary. If the total Hamiltonian  $H(t)$  has an explicit time dependence, then this dependence will generate an additional phase evolution. For example, precision spectroscopy of trapped ions must contend with the fact that the ions are in motion, and experience varying ambient fields during the course of their orbit. Likewise, ultracold atomic samples confined in an optical dipole trap experience, in principle, an oscillating electric field due to the laser field that provides the trapping potential.

In the case of a Hamiltonian with a slow, periodic time dependence  $H(t+\tau) = H(t)$ , Berry [1] has given a famous description of the additional phase. Berry's original treatment requires that the period  $\tau$  be far larger than any other relevant time scale of the system, and thus finds an "adiabatic" phase shift. This shift is largely independent of the detailed way in which the Hamiltonian varies with time, and leads to an elegant geometric description of the phase [1, 2, 3, 4, 5, 6, 7]. Extensions to this formalism have considered the next-order corrections if the rate of change of the Hamiltonian is not strictly adiabatic [8, 9, 10, 11, 12, 13, 14]. A more general Floquet theory has also been advanced, which allows one to consider the effect of overtones of the fundamental period  $\tau$  [14, 15, 16]. In addition, the ideas have been extended to particles with dynamic properties [17], gauge structure [18], the Quantum Hall Effect [19], and to relativistic effects using the Dirac equation [20].

Thus far, applications of the Berry phase have mostly considered the effect of the time-dependence on quantum mechanical particles without internal structure, although atoms with two or several levels have been considered [16, 21, 22]. However, the job of precision spectroscopy is precisely to reveal this internal structure. Corrections to Berry's phase arising from degrees of freedom internal to an atom or molecule is our concern in this article. To establish a concrete formalism for this, we will consider a particular case, namely, a diamagnetic or electrically polar species in the presence of a magnetic or electric field, whose

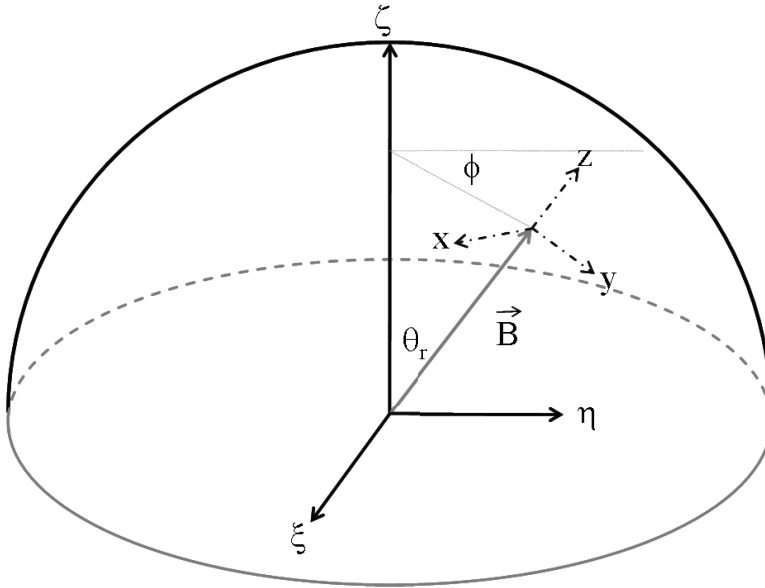


FIG. 1: The axis of rotation with laboratory-fixed coordinates  $\{\xi, \eta, \zeta\}$  as well as the field coordinates defined by  $\{x, y, z\}$ . The field direction rotates about the  $\zeta$ -axis with angular frequency  $\omega_r$ .

direction precesses on a cone with an angular frequency  $\omega_r$  (see Fig. I). The system evolves in time according to the field variation, combined with whatever intrinsic Hamiltonian governs the particle's internal structure. The internal structure dictates regimes of linear and quadratic Zeeman (Stark) shifts with respect to the applied magnetic (electric) field.

A main point in deriving the non-dynamic phase in this situation is to recognize the periodicity of the driving field. By analogy to the periodic driving of a near-resonant laser field applied to a two-level atom, we consider “field-dressed” states of the Hamiltonian in the spirit of quantum optics [22, 23]. This viewpoint effectively counts the energy of the atom itself, plus that of the photons of frequency  $\omega_r$  arising from the driving field. The additional energy shift due to the rotating field is then equivalent to the ac Stark effect in optics. By constructing the complete Hamiltonian in this way, we are able to accommodate the particle's internal structure. We are also able to consider arbitrary rates of rotation, not just those that are adiabatic with respect to the particle's Hamiltonian. Nevertheless, in what follows we will focus primarily on results for low rotation rates, to better draw analogies with the usual adiabatic phase.

This article is structured as follows. In Sec. II we work out the general transformation

from a Hamiltonian with an explicit time-rotating field, into an effective dressed Hamiltonian whose eigen-energies yield the shifted energies. We illustrate this transformation first with a simple two-level atom, then generalize it to an arbitrary atom or molecule. In Sec. III we briefly re-visit a structureless particle with arbitrary total spin, showing that our results reduce to Berry's in the limit of slow rotation. Sec. IV illustrates the application of the method to a particle composed of two interacting spin-1/2 objects, to show most clearly the effect of their fine structure on the Berry phase. Finally, Sec. V considers a simple case of a dipolar molecule in a rotating electric field, to assess the influence of molecular end-over-end rotation on the phase.

## II. DERIVATION OF THE DRESSED STATES

This section lays the ground work for the formalism. It is composed of two subsections. The first works out the derivation for a spin-1/2 system in a rotating magnetic field, and will illustrate simply and cleanly the basic idea. The second subsection contains a generalization of this same derivation to explicitly account for an arbitrary quantum mechanical object with total spin  $j$ .

### A. Spin-1/2 interaction

The most elementary of quantum mechanical objects is the spin-1/2 particle. Its interaction with a time-dependent magnetic field is

$$H(t) = -\vec{\mu}_m \cdot \vec{\mathcal{B}}(t). \quad (1)$$

There are two coordinate systems that we will find useful, the lab-fixed Cartesian coordinates labeled  $(\xi, \eta, \zeta)$ ; and a coordinate system  $(x, y, z)$  rotating with the magnetic field, where  $\hat{z} = \hat{\mathcal{B}}$  (see Fig. I). In the lab-fixed frame the field rotation is given by its components

$$\begin{aligned} \vec{\mathcal{B}}(t) &= (\mathcal{B}_\xi(t), \mathcal{B}_\eta(t), \mathcal{B}_\zeta(t)) \\ &= \mathcal{B} (\sin \theta_r \cos \omega_r t, \sin \theta_r \sin \omega_r t, \cos \theta_r), \end{aligned} \quad (2)$$

where  $\phi = \omega_r t$ . The field makes an angle  $\theta_r$  with respect to the lab-fixed  $\zeta$  axis, which is also the axis it rotates about. In other words we identify a vector  $\vec{\omega}_r = \omega_r \hat{\zeta}$ , the axis defining the field's rotation.

The spin-field interaction has its own characteristic frequency,

$$-\vec{\mu}_m \cdot \vec{\mathcal{B}} = \frac{g\mu_B \mathcal{B}}{2} \vec{\sigma} \cdot \hat{\mathcal{B}} \equiv \frac{\hbar}{2} \omega_L \vec{\sigma} \cdot \hat{B}, \quad (3)$$

in terms of the Pauli matrices  $\vec{\sigma}$  and the ( $m$ -independent) Larmor frequency  $\omega_L = g\mu_B \mathcal{B}$ .  $g$  is the usual  $g$ -factor; e.g.  $g \approx 2.0023$  for the simple case of the spin-1/2 electron. The eigen-energies of the non-rotating spin-field interaction are  $\pm \hbar \omega_L/2$ . By causing the field to rotate, we expect to find apparent energies that are different from these. From this point forward we shall work in atomic units and set  $\hbar = 1$ .

In this simple example, we can easily cast the Hamiltonian in matrix form, using the basis of spin functions  $|m_\zeta = \pm 1/2\rangle$ , with quantization along the  $\zeta$ -axis. These states themselves are then independent of time, which would not be the case if we used the eigenstates referred to the (moving) magnetic field. Using the explicit forms for the Pauli matrices, we get

$$H(t) = \frac{\omega_L}{2} \begin{pmatrix} \cos(\theta_r) & \sin(\theta_r) e^{-i\omega_r t} \\ \sin(\theta_r) e^{i\omega_r t} & -\cos(\theta_r) \end{pmatrix}. \quad (4)$$

While this basis seems cumbersome it is quite useful for the dressed-state calculation we wish to perform.

We begin in the most general way by writing an *ansatz* wave function in this basis:

$$|\psi(t)\rangle = \begin{pmatrix} \alpha(t) e^{-i\omega_+ t} \\ \beta(t) e^{-i\omega_- t} \end{pmatrix}, \quad (5)$$

where we leave the frequencies  $\omega_\pm$  unspecified for now. We note a very similar development has recently been carried out by the authors of Ref. [24] for a two-level system. The constants will be chosen later to cancel any time dependence in the system. The wave function  $|\psi\rangle$  satisfies the time-dependent Schrödinger equation (TDSE)

$$i \frac{\partial |\psi\rangle}{\partial t} = H(t) |\psi\rangle. \quad (6)$$

Inserting the *ansatz* (5) into the TDSE yields the equation of motion for  $\alpha$  and  $\beta$ . This new equation introduces terms which amount to a kind of effective Hamiltonian due to rotation. Moving these to the RHS of the TDSE, we get

$$i \frac{\partial}{\partial t} \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = \begin{pmatrix} +\frac{\omega_L}{2} \cos \theta_r - \omega_+ & \frac{\omega_L}{2} \sin \theta_r e^{-i(\omega_r + \omega_- - \omega_+)t} \\ \frac{\omega_L}{2} \sin \theta_r e^{i(\omega_r + \omega_- - \omega_+)t} & -\frac{\omega_L}{2} \cos \theta_r - \omega_- \end{pmatrix}. \quad (7)$$

Formally, this is still a time-dependent Hamiltonian unless we use the freedom in choosing the frequencies  $\omega_{\pm}$  to get rid of this dependence. We could achieve this by setting

$$\omega_r + \omega_- = \omega_+. \quad (8)$$

There are many ways to accomplish this, but a particularly symmetric and appealing one is to let

$$\omega_{\pm} = \pm \frac{\omega_r}{2}, \quad (9)$$

or, even more to the point,

$$\omega_{m_{\zeta}} = m_{\zeta} \omega_r \quad (10)$$

for each angular momentum projection  $m_{\zeta}$  along the rotation axis. This last statement is very general, and will motivate our choice of wave function in the spin- $j$  case below.

In any event, we are presented with a formally time-independent Hamiltonian, dressed by the rotation:

$$H_{\text{dressed}} = H_{\text{nr}} + H_{\text{r}}, \quad (11)$$

where the magnetic-field part, in the laboratory-fixed  $|m_{\zeta}\rangle$  basis, looks like

$$H_{\text{nr}} = \frac{\omega_L}{2} \begin{pmatrix} \cos(\theta_r) & \sin(\theta_r) \\ \sin(\theta_r) & -\cos(\theta_r) \end{pmatrix}, \quad (12)$$

and is exactly the same as having the magnetic field tilted at an angle  $\theta_r$  with respect to the quantization axis in the direction of the  $x$ -axis, as specified by  $\phi = \omega_r t = 0$ . Rather, the vestiges of rotation show up in the effective term

$$H_{\text{r}} = \frac{\omega_r}{2} \begin{pmatrix} -1 & 0 \\ 0 & +1 \end{pmatrix} = -\text{diag}(m_{\zeta} \omega_r). \quad (13)$$

Now that we have a time-independent Hamiltonian we write

$$\begin{pmatrix} \alpha(t) \\ \beta(t) \end{pmatrix} = \begin{pmatrix} \alpha_0 \\ \beta_0 \end{pmatrix} \exp(-i\lambda t), \quad (14)$$

where  $\lambda$  is the effective, or dressed, eigen-frequency of the “stationary” state defined by  $H_{\text{nr}} + H_{\text{r}}$ .  $\lambda$  is, in other words, the eigenvalue of  $H_{\text{dressed}}$ :

$$\lambda_{\pm} = \pm \frac{1}{2} \sqrt{\omega_L^2 - 2\omega_L \omega_r \cos(\theta_r) + \omega_r^2}. \quad (15)$$

Thus the apparent energy difference, measured by the phase difference accumulated during a hold time  $\tau$ , would be  $(\lambda_+ - \lambda_-)\tau = \phi_+ - \phi_-$ , rather than the  $(\omega_{+1/2} - \omega_{-1/2})\tau = \varphi_+ - \varphi_-$  that would measure the intrinsic energy splitting. The difference between these two we attribute to a Berry-like phase in each state defined as (where we assume the dynamical phase is unaffected by the rotation)

$$\gamma_{\pm} = \phi_{\pm} - \varphi_{\pm}, \quad (16)$$

where  $\phi$  ( $\varphi$ ) is the total (dynamic) phase of the system. This removes the dynamical phase  $\varphi$  that would have been accumulated in the absence of rotation. Hence  $\gamma/\tau$  represents the error introduced by the field's rotation into a measurement of the state's energy.

The dressed eigen-energies are actually independent of whether or not the time  $\tau$  refers to one period of the rotating field. To connect explicitly to Berry's phase, let  $\tau = 2\pi/\omega_r$  be one period of the field's rotation. Berry's phase results when the field rotation is slow, i.e. when the frequency of the rotating field is small compared to the Larmor precession frequency of the spin. If we assert that  $\omega_r \ll \omega_L$ , then the spin is expected to precess around the instantaneous field direction, and to follow this direction during the rotation of the field. In this limit the dressed-state eigenvalue of one state, say  $\lambda_+$ , is

$$\lambda_+ \approx \frac{\omega_L}{2} - \frac{\omega_r}{2} \cos(\theta_r). \quad (17)$$

In a period  $\tau$  the additional phase accumulated by this state is (see (16))

$$\begin{aligned} \gamma_+^{(0)} &\approx -\frac{1}{2}\omega_r\tau \cos(\theta_r) \\ &\approx \pi - \frac{1}{2}(2\pi) \cos(\theta_r). \end{aligned} \quad (18)$$

Here we have used the freedom to add  $\pi$  to the phase, which amounts to an unobservable overall change of sign of the dressed eigenstate ( $\alpha_0 \beta_0$ ).

The ordinary dynamical phase  $\varphi = \omega_L\tau$  is canceled out in (18) leaving only the terms due to the slow rotation of the field. This remainder can be written

$$\gamma^{(0)} = \frac{1}{2}(2\pi)(1 - \cos(\theta_r)) = \frac{1}{2}\Delta\Omega \Rightarrow m\Delta\Omega, \quad (19)$$

where

$$\Delta\Omega = \int_0^{2\pi} d\phi \int_0^{\theta_r} \sin(\theta) d\theta = 2\pi(1 - \cos(\theta_r)) \quad (20)$$

is the solid angle subtended by the vector describing the direction of the rotating field. In other words, this additional phase  $\gamma^{(0)}$  is exactly Berry's phase  $m\Delta\Omega$ , for a particle with spin projection  $m = 1/2$  along the  $\hat{\zeta}$ -axis. Similarly, for the  $m = -1/2$  state there arises an additional phase  $-(1/2)\Delta\Omega$ .

We have cast the Hamiltonian  $H$  in (11) in the basis where angular momentum projection along the rotation axis  $\hat{\omega}_r = \hat{\zeta}$  is a good quantum number. Yet, in the limit of slow rotation that is particularly important, the relevant states are those where  $m$  is quantized along the field axis. We should therefore cast  $H$  in the basis of states  $|s m\rangle$  quantized along the field axis, which we call the  $\hat{z}$ -axis (see Fig. 1).

Using the *ansatz* (5), our dressed Hamiltonian represents a Hamiltonian  $H_{\text{nr}}$  that is already diagonal in this basis, and reads

$$H_{\text{nr}} = \frac{\omega_L}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (21)$$

To cast the rotational piece  $H_r$  in this basis, we make explicit the notation  $|s m_\zeta\rangle$  for spin states quantized along the rotation axis  $\hat{\zeta}$ ; and  $|s m\rangle$  for spin states quantized along the field axis  $\hat{z}$ . The rotation from  $\hat{\zeta}$  to  $\hat{z}$  is accomplished through the Euler angles  $R = (0, \theta_r, 0)$ . The transformation from  $|s m_\zeta\rangle$  to  $|s m\rangle$  is given by [25]

$$|s m\rangle = D(R)|s m_\zeta\rangle \quad (22)$$

$$\begin{aligned} &= \sum_{m'_\zeta} |s m'_\zeta\rangle \langle s m'_\zeta| D(R) |s m_\zeta\rangle \\ &= \sum_{m'_\zeta} |s m'_\zeta\rangle D_{m'_\zeta m_\zeta}^s(R). \end{aligned} \quad (23)$$

The Wigner rotation matrices have a simple explicit form:

$$D_{m'_\zeta m_\zeta}^s(0, \theta_r, 0) = e^{-im'_\zeta 0} d_{m'_\zeta m_\zeta}^s(\theta_r) e^{-im_\zeta 0}, \quad (24)$$

where

$$d_{1/2, 1/2}^{1/2} = d_{-1/2, -1/2}^{1/2} = \cos\left(\frac{\theta_r}{2}\right) \quad (25)$$

$$d_{-1/2, 1/2}^{1/2} = -d_{1/2, -1/2}^{1/2} = \sin\left(\frac{\theta_r}{2}\right). \quad (26)$$



Using the fact that  $H_r$  is diagonal in the rotation basis with values  $-m_\zeta\omega_r$ , it is easily verified that, in the rotating-frame basis,

$$H_r = \frac{\omega_r}{2} \begin{pmatrix} -\cos(\theta_r) & -\sin(\theta_r) \\ -\sin(\theta_r) & \cos(\theta_r) \end{pmatrix}. \quad (27)$$

Therefore, the Hamiltonian matrix referred to the field axis is

$$H = H_{\text{nr}} + H_r \quad (28)$$

$$= \begin{pmatrix} \frac{\omega_L}{2} - \frac{\omega_r}{2} \cos \theta_r & -\frac{\omega_r}{2} \sin \theta_r \\ -\frac{\omega_r}{2} \sin \theta_r & -\frac{\omega_L}{2} + \frac{\omega_r}{2} \cos \theta_r \end{pmatrix}. \quad (29)$$

And this matrix gives has the same eigenvalues as (15), as is expected when one merely performs a unitary transformation on the system.

In this basis, the Hamiltonian is already diagonal in the absence of rotation. Thus for a small rotation it is nearly diagonal, and the eigenvalues are easily estimated in perturbation theory. Indeed, Berry's energy follows immediately from the diagonal perturbations in this matrix: it is  $-(\omega_r/2)\cos(\theta_r)$  for  $m = +1/2$ , and  $+(\omega_r/2)\cos(\theta_r)$  for  $m = -1/2$ .

To summarize, for a Hamiltonian of the form  $H(t)$  that we have been dealing with, the rotation-dressed energies are given by the eigenvalues of the time-independent operator

$$H_{\text{dressed}} = H(t=0) - \omega_r s_\zeta = H(t=0) - \omega_r (\cos(\theta_r)s_z - \sin(\theta_r)s_x). \quad (30)$$

For more general systems incorporating internal structure, all that is required is to include the appropriate structure in  $H(t=0)$ , as we will now see.

## B. A general spin- $j$ system

Having provided the groundwork by working out the simple spin-1/2 system, we now proceed with a general derivation for the spin- $j$  system. A system such as this can be described by the following Hamiltonian

$$H(t) = H_0 - \vec{\mu} \cdot \vec{\mathcal{F}}(t), \quad (31)$$

where  $\vec{\mathcal{F}}(t)$  is an external field, electric or magnetic, that acts on an appropriate moment  $\vec{\mu}$  of the atom or molecule.  $\vec{\mathcal{F}}$  rotates on a cone at frequency  $\omega_r$  and tilt angle  $\theta_r$  just as  $\vec{B}$  did

in the previous section and depicted in Fig. I. Here  $H_0$  is a Hamiltonian in the absence of the applied rotating field. It can be used to describe the hyperfine elements of an atom or it can be a detailed molecular Hamiltonian that includes such items as rotation, spin-spin, nuclear spin, or lambda doubling. The Hamiltonian can equally be represented in a basis referred to the axis of rotation or to the instantaneous field axis. Later we will take the instantaneous field axis, just as we did above. We will, as in the previous section, begin by quantizing along the axis of rotation. We take this structured object and place it in a rotating field  $\vec{\mathcal{F}}$ ; this can be either electric or magnetic provided there is an electric or magnetic dipole that interacts with the field in the usual way, i.e. it is a scalar interaction of two vectors.

To work with this Hamiltonian, it is again convenient to pick two basis sets:

$$\begin{aligned} &|(\kappa)jm_\zeta\rangle \\ &|(\kappa)jm_j\rangle \end{aligned} \tag{32}$$

Because  $j$  is the total of all relevant angular momenta, its projection onto an axis is unambiguously defined as  $m_\zeta$  in the lab frame and  $m_j$  in the rotating frame, as above. Here  $\kappa$  is a shorthand notation for all the other quantum numbers required to specify the state.

To deal with the explicit time dependence of the field rotation, we will expand into the lab basis first, and will make the same trial wave function that was motivated above;

$$|\psi(t)\rangle = \sum_{\kappa',j',m'_\zeta} C_{\kappa',j',m'_\zeta} e^{-im'_\zeta\omega_r t} |(\kappa')j'm'_\zeta\rangle. \tag{33}$$

We have explicitly included a time dependent phase factor with phase  $m_\zeta\omega_r$ , which is akin to the spin-1/2 case, cf. (10). Taking the time derivative for the TDSE and projecting onto a particular state, gives

$$\langle(\kappa)jm_\zeta|i\frac{d|\psi\rangle}{dt} = \left(i\dot{C}_{\kappa,j,m_\zeta} + m_\zeta\omega_r C_{\kappa,j,m_\zeta}\right) e^{-im_\zeta\omega_r t}. \tag{34}$$

As for the internal Hamiltonian  $H_0$ , it may or may not be diagonal in our basis, but it does not depend on any external field. Therefore it can be represented in a basis where it is diagonal in  $m_\zeta$ , whereby

$$\langle(\kappa)jm_\zeta|H_0|\psi\rangle = \sum_{\kappa',j',m'_\zeta} e^{i(m_\zeta-m'_\zeta)\omega_r t} \langle(\kappa)jm_\zeta|H_0|(\kappa')j'm'_\zeta\rangle C_{\kappa',j',m'_\zeta} \delta_{m_\zeta,m'_\zeta} \tag{35}$$

To treat the field interaction, we use the language of tensor algebra, and express the spherical components of  $\vec{\mathcal{F}}$  in the lab frame as an explicit rotation from  $\vec{\mathcal{F}}$  in the rotating frame (whose  $z$  axis is, of course, defined by the instantaneous direction of  $\vec{\mathcal{F}}$  itself):

$$\begin{aligned}\mathcal{F}_\iota &= \sum_q \mathcal{F}_q \mathcal{D}_{\iota q}^{1*}(\omega_r t, \theta_r, 0) \\ &= \mathcal{F} \mathcal{D}_{\iota 0}^{1*}(\omega_r t, \theta_r, 0).\end{aligned}\tag{36}$$

$\mathcal{F}$  is the magnitude of the field, and  $q$  is its spherical projection in the rotating frame. But  $\vec{\mathcal{F}}$  defines this frame so only the values of  $q = 0$  will contribute.  $\mathcal{D}$  is a Wigner rotation matrix. In a similar manner, the dipole moment  $\vec{\mu}$  is determined by its spherical components such that

$$\begin{aligned}-\vec{\mu} \cdot \vec{\mathcal{F}} &= -\sum_\iota (-1)^\iota \mu_\iota \mathcal{F}_{-\iota} \\ &= -\mathcal{F} \sum_\iota (-1)^\iota \mu_\iota \mathcal{D}_{-\iota 0}^{1*}(\omega_r t, \theta_r, 0) \\ &= -\mathcal{F} \sum_\iota \mu_\iota e^{-i\omega_r t} d_{-\iota 0}^1(\theta_r).\end{aligned}\tag{37}$$

This uses the explicit expression for  $\mathcal{D}$  in terms of a little- $d$  function [25].

Just as we treated the internal degrees of freedom in  $H_0$  we must now treat the field interaction.

$$\begin{aligned}\langle (\kappa) j m_\zeta | -\vec{\mu} \cdot \vec{\mathcal{F}} | \psi \rangle &= -\mathcal{F} \sum_{\kappa' j' m'_\zeta \iota} (-1)^\iota \langle (\kappa) j m_\zeta | \mu_\iota | (\kappa') j' m'_\zeta \rangle \times \\ &\quad C_{\kappa', j', m'_\zeta} d_{-\iota 0}^1(\theta_r) e^{i\omega_r t(-\iota - m'_\zeta)}.\end{aligned}\tag{38}$$

Piecing together the different parts, and multiplying through by  $e^{im_\zeta \omega_r t}$ , we arrive at the TDSE for the coefficients  $C$ :

$$\begin{aligned}i\dot{C}_{\kappa, j, m_\zeta} + \omega_r m_\zeta C_{\kappa, j, m_\zeta} &= \sum_{\kappa' j'} \langle (\kappa) j m_\zeta | H_0 | (\kappa') j' m_\zeta \rangle C_{\kappa', j', m_\zeta} - \\ &\quad \mathcal{F} \sum_{\kappa' j' m'_\zeta \iota} (-1)^\iota \langle (\kappa) j m_\zeta | \mu_\iota | (\kappa') j' m'_\zeta \rangle C_{\kappa', j', m'_\zeta} \times \\ &\quad d_{-\iota 0}^1(\theta_r) e^{i\omega_r t(-\iota - m'_\zeta + m_\zeta)}.\end{aligned}\tag{39}$$

Now, we have not specified what field  $\vec{\mathcal{F}}$  is, nor which structural degrees of freedom are involved in making the dipole  $\vec{\mu}$ , and it does not matter. All that matters is that  $\vec{\mu}$  is

a vector, in which case the Wigner-Eckhart theorem applies [25]. In the total angular momentum basis, we must have

$$\langle(\nu)jm_\zeta|\mu_\iota|(\nu')j'm'_\zeta\rangle \propto \begin{pmatrix} j & 1 & j' \\ -m_\zeta & \iota & m'_\zeta \end{pmatrix}, \quad (40)$$

where the proportionality constant involves the reduced matrix element. Then the conservation of angular momentum implies that  $m'_\zeta - m_\zeta = -\iota$ . However, this immediately removes the time-dependence in the exponential term in (39). In fact, as alluded to earlier, this statement says that any angular momentum imparted by the rotating field must be accounted for in the projection  $m_\zeta$ .

In some cases it will prove more useful to keep track of the individual spin components  $m_{\zeta i}$  separately. For instance, suppose there were two angular momenta,  $m_{\zeta 1}$  and  $m_{\zeta 2}$ : we would have two projection terms that would each evolve as  $e^{i m_{\zeta 1} \omega_r t}$  and  $e^{i m_{\zeta 2} \omega_r t}$ . Terms in the Hamiltonian which describe the interaction of the two spins are of the form  $\vec{s}_1 \cdot \vec{s}_2$  for which we find scales as

$$\begin{aligned} & \langle s_1 m_{\zeta 1} s_2 m_{\zeta 2} | \vec{s}_1 \cdot \vec{s}_2 | s'_1 m'_{\zeta 1} s'_2 m'_{\zeta 2} \rangle \propto \\ & \begin{pmatrix} s_1 & 1 & s'_1 \\ -m_{\zeta 1} & p & m'_{\zeta 1} \end{pmatrix} \begin{pmatrix} s_2 & 1 & s'_2 \\ -m_{\zeta 2} & -p & m'_{\zeta 2} \end{pmatrix} e^{i(m_{\zeta 1} - m'_{\zeta 1})\omega_r t} e^{i(m_{\zeta 2} - m'_{\zeta 2})\omega_r t}. \end{aligned} \quad (41)$$

The proportionality involves a reduced matrix element. By the conservation of angular momentum we find that  $m_{\zeta 1} - m'_{\zeta 1} = -(m_{\zeta 2} - m'_{\zeta 2})$  and the phase factor is still canceled out. In fact, for any such interaction between two spins, the conservation of angular momentum forces the time dependence to cancel out.

With the time-dependence removed, Eq. (39) reduces to the Schrödinger equation for a non-rotating field tilted at an angle  $\theta_r$  from the rotation axis, just as in the spin-1/2 example in the prior subsection. This introduces an additional term on the left of (39), which is moved to the RHS and interpreted as an effective Hamiltonian. Thus if  $H_0$  is presented in the basis  $|(\kappa)j m_j\rangle$  diagonal with respect to the field, then the matrix to be diagonalized is

$$\begin{aligned} H_{\text{dressed}} &= H_0 - \vec{\mu} \cdot \vec{\mathcal{F}} - \omega_r m_\zeta \\ &= H_0 - \vec{\mu} \cdot \vec{\mathcal{F}} - \omega_r j_\zeta, \end{aligned} \quad (42)$$

where  $m_\zeta$  is the eigenvalue of the  $j_\zeta$  operator. As before, we can rotate this Hamiltonian from the  $m_\zeta$  basis to the  $m_j$  basis. Since  $H_0$  does not depend on either  $m_\zeta$  or  $m_j$ , it is

unaffected by this rotation. In the frame of the instantaneous field where  $m_j$  is the good quantum number, we can write the dressed Hamiltonian as

$$\begin{aligned} H_{\text{dressed}} &= H_0 - \vec{\mu} \cdot \vec{\mathcal{F}} - \hat{\omega}_r \cdot \vec{j}, \\ &= H_0 - \vec{\mu} \cdot \vec{\mathcal{F}} - \omega_r (\cos(\theta_r) j_z - \sin(\theta_r) j_x). \end{aligned} \quad (43)$$

This dressed Hamiltonian is the main result of this article. This Hamiltonian has been previously formulated in NMR studies [26]. In the following sections we will apply it to several elementary cases of interest.

### III. PURE SPIN- $s$ SYSTEM

As the simplest application of the general method beyond the spin-1/2 particle, we consider in this section a structureless particle of arbitrary spin  $s$ , as was considered in the original formulation of Berry [1]. This spin interacts with a magnetic field that rotates at an angle  $\theta_r$  with respect to the axis of rotation. Using the result from (43), this system is described by the Hamiltonian

$$H_{\text{dressed}} = \omega_L s_z - \omega_r (\cos(\theta_r) s_z - \sin(\theta_r) s_x), \quad (44)$$

where  $\omega_L = g_s \mu_B \mathcal{B}$  is the  $m$ -independent Larmor precession frequency and  $g_s$  is the  $g$ -factor for the spin- $s$ . For this section, we have reverted to the usual notation  $s$  and  $m$  for the spin and its projection onto the instantaneous field axis.

For this structureless particle, the Hamiltonian (44) is represented by a  $(2s+1) \times (2s+1)$  tridiagonal matrix, in the basis of states  $|s m\rangle$ . This matrix is explicitly given by

$$H = \begin{pmatrix} m a & b(s, m) & 0 & 0 & \dots \\ b(s, m) & (m-1) a & b(s, m-1) & 0 & \vdots \\ 0 & b(s, m-1) & \ddots & \ddots & b(s, -m+1) \\ \vdots & 0 & b(s, -m+1) & -(m-1) a & b(s, -m) \\ \dots & 0 & 0 & b(s, -m) & -m a. \end{pmatrix}, \quad (45)$$

where  $a = \omega_L - \omega_r \cos(\theta_r)$  and  $b(s, m) = (1/2) \sqrt{s(s+1) - m(m-1)} \omega_r \sin(\theta_r)$ . Appendix A sketches a derivation of the eigenvalues of this matrix, which are

$$\lambda_m = m \sqrt{\omega_L^2 + \omega_r^2 - 2\omega_r \omega_L \cos(\theta_r)}, \quad (46)$$

where  $m$  takes on the values  $-s, \dots, +s$  in integer steps.

The usual Berry phase is obtained in the adiabatic limit where  $\omega_r \ll \omega_L$ , in which case

$$\lambda_m \approx m\omega_L - m\omega_r \cos(\theta_r). \quad (47)$$

The magnetic field completes one rotation in a time  $\tau = 2\pi/\omega_r$ . In this time the spin accumulates a dynamical phase  $\varphi_m = m\omega_L\tau$ . Beyond this, it acquires an additional phase  $\gamma_m = \phi_m - \varphi_m$ , where  $\phi_m = \lambda_m\tau$ , given to lowest order by

$$\gamma_m^{(0)} = -m2\pi \cos(\theta_r) \Rightarrow 2\pi m(1 - \cos(\theta_r)). \quad (48)$$

In the final step, we use the fact that adding  $2\pi m$  (where  $m$  is either integer or half-integer) amounts to adding an unobservable phase of  $\pm 1$  to the system. We see that the phase  $\gamma_m^{(0)}$  accumulated is exactly that given by the result of Berry;  $m$  times the solid angle subtended by the rotation. We can also extend this solution to regimes of non-adiabaticity. The first order correction in  $\omega_r/\omega_L$  is

$$\gamma_m^{(1)} = 2\pi m \frac{\omega_r}{2\omega_L} \sin^2(\theta_r), \quad (49)$$

which has already been identified elsewhere [8, 14]. Based on our explicit formula, we can extract corrections to any desired order, at least for fields undergoing the simple motion in Fig. I. This additional phase can be expanded to any desired order in the adiabatic parameter  $\omega_r/\omega_L$ . For example, the second and third-order  $\gamma^{(k)}$  corrections are

$$\gamma_m^{(2)} = 2\pi m \frac{\omega_r^2}{2\omega_L^2} \cos(\theta_r) \sin^2(\theta_r) \quad (50)$$

$$\gamma_m^{(3)} = 2\pi m \frac{\omega_r^3}{16\omega_L^3} (3 + 5 \cos(2\theta_r)) \sin^2(\theta_r) \quad (51)$$

Using the general dressed formalism, the limit of fast field rotation can also be described. The phase  $\gamma_m$  (after one field period) can be approximated in this limit ( $\omega_r \gg \omega_L$ ):

$$\gamma_m \approx 2\pi m - 2\pi m \frac{\omega_L}{\omega_r} \cos(\theta_r), \quad (52)$$

and the first term is unobservable. In this case the dominant energy, as manifested in the phase, is the photon energy due to the time-periodic field. On top of this, the magnetic field interaction itself makes a small correction. This is clearly not the appropriate limit in which to perform precision spectroscopy, since small uncertainties in the field rotation rate would dominate the observable Larmor frequency.

The structureless spin problem can be solved analytically for arbitrary rotation rates since the spin precesses about an *effective* magnetic field in the rotating coordinate system [27]. The effective magnetic field makes an angle with respect to the positive  $\zeta$ -axis,  $\theta_r^*$ , satisfying

$$\begin{aligned}\sin(\theta_r^*) &= \frac{\sin(\theta_r)}{\sqrt{1 - 2\frac{\omega_r}{\omega_L} \cos(\theta_r) + \left(\frac{\omega_r}{\omega_L}\right)^2}}, \\ \cos(\theta_r^*) &= \frac{\cos(\theta_r) - \frac{\omega_r}{\omega_L}}{\sqrt{1 - 2\frac{\omega_r}{\omega_L} \cos(\theta_r) + \left(\frac{\omega_r}{\omega_L}\right)^2}}.\end{aligned}\tag{53}$$

This angle smoothly transitions from  $\theta_r^* \approx \theta_r$  for  $\omega_r \ll \omega_L$  to  $\theta_r^* \approx \pi$  for  $\omega_r \gg \omega_L$ . Equivalently, the angular deviation,  $\Delta\theta_r = \theta_r^* - \theta_r$ , between the effective magnetic field and the true magnetic field can be described through

$$\begin{aligned}\sin(\Delta\theta_r) &= \frac{\frac{\omega_r}{\omega_L} \sin(\theta_r)}{\sqrt{1 - 2\frac{\omega_r}{\omega_L} \cos(\theta_r) + \left(\frac{\omega_r}{\omega_L}\right)^2}}, \\ \cos(\Delta\theta_r) &= \frac{1 - \frac{\omega_r}{\omega_L} \cos(\theta_r)}{\sqrt{1 - 2\frac{\omega_r}{\omega_L} \cos(\theta_r) + \left(\frac{\omega_r}{\omega_L}\right)^2}},\end{aligned}\tag{54}$$

where this deviation smoothly transitions from  $\Delta\theta_r^* \approx 0$  for  $\omega_r \ll \omega_L$  to  $\Delta\theta_r^* \approx \pi - \theta_r$  for  $\omega_r \gg \omega_L$ .

The total phase shift,  $\phi_m = \varphi_m^* + \gamma_m^*$ , accumulated during one revolution of the magnetic field can be broken up into a dynamic contribution,  $\varphi_m^*$ , and a geometric contribution,  $\gamma_m^*$ , where [26, 28, 29]

$$\begin{aligned}\varphi_m^* &= 2\pi m \frac{\omega_L}{\omega_r} \cos(\Delta\theta_r), \\ \gamma_m^* &= 2\pi m (1 - \cos(\theta_r^*)).\end{aligned}\tag{55}$$

$\varphi_m^*$  and  $\gamma_m^*$  are slightly different than the phases defined in Eq. (16). In this definition, both the dynamical phase and the geometric phase acquire non-adiabatic corrections. In the limit of very slow rotation  $\omega_r$ , we have that  $\varphi_m^* \rightarrow \varphi_m$ . Grouping terms together and simplifying provides the total accumulated phase  $\phi_m$ :

$$\begin{aligned}\phi_m &= 2\pi m \left( 1 + \frac{\omega_L}{\omega_r} \sqrt{1 - 2\frac{\omega_r}{\omega_L} \cos(\theta_r) + \left(\frac{\omega_r}{\omega_L}\right)^2} \right), \\ &= 2\pi m \left( 1 + \sqrt{1 - 2\frac{\omega_L}{\omega_r} \cos(\theta_r) + \left(\frac{\omega_L}{\omega_r}\right)^2} \right).\end{aligned}\tag{56}$$

The total phase shift arrived at in this geometrical way is the same as the dressed state derivation —  $(2\pi/\omega_r)\lambda_m$ , where  $\lambda_m$  is defined in (46) — apart from a factor of  $2\pi m$ , which is unobservable for integer or half-integer values of  $m$ . This is a semi-classical geometric procedure that yields the non-adiabatic corrections to Berry's result.

For the above calculations, we have taken  $\vec{\mu} = -g_s\mu_B\vec{S}$  and  $\omega_r$  to have a *positive* sense about the  $\zeta$ -axis. For the single revolution phase shifts,  $\gamma_m^{(i)}$ , terms proportional to *odd* powers of  $\omega_r/\omega_L$  change sign when the g-factor changes sign, while terms proportional to *even* powers of  $\omega_r/\omega_L$  change sign when the sense of rotation changes sign. For the expressions defining the angle,  $\theta_r^*$ , and angular deviation,  $\Delta\theta_r$ , changing the sign of the g-factor (sense of rotation) directly changes the sign of  $\omega_L$  ( $\omega_r$ ).

A spectroscopic measurement would involve finding the energy difference between two states with different values of  $m$ , with difference  $\Delta m$ . In a Ramsey-type experiment, this measurement seeks to measure the phase difference  $\omega_L\Delta m\tau = \Delta\varphi$ . In a rotating field, however, the experiment will produce a measurement of  $\Delta\phi = \Delta m\sqrt{\omega_L^2 + \omega_r^2 - 2\omega_L\omega_r\cos(\theta_r)}\tau$ , and thus will introduce an error. This error is given by the difference  $\Delta\gamma = \gamma_m - \gamma_{m'} = \Delta\phi - \Delta\varphi$ , and is plotted in Fig. III as a function of rotation rate. The different curves represent different values of the tilt angle  $\theta_r$ .

#### IV. STRUCTURED SPIN- $J$ SYSTEM

More generally, atoms and molecules are composite objects made of individual spins, which are moreover coupled together to create fine or hyperfine structure. For example, alkali atoms couple the electronic and nuclear spins into a total hyperfine state. The resulting angular momentum structure will have a bearing on the non-adiabatic corrections to the geometric phase accumulated.

As a simple illustration of our formalism, we consider a composite particle composed of two spin-1/2 objects. This example goes beyond the structureless particle often envisioned by the usual Berry theory. The dressed Hamiltonian is given by

$$H_{\text{dressed}} = \omega_1 j_{1z} + \omega_2 j_{2z} + \Delta \vec{j}_1 \cdot \vec{j}_2 - \omega_r (\cos(\theta_r) J_z - \sin(\theta_r) J_x), \quad (57)$$

where  $\omega_i = g_i\mu_B\mathcal{B}$  is the Larmor precession frequency of spin  $j_i$ ;  $\vec{J}$  is the vector sum of  $\vec{j}_1$  and  $\vec{j}_2$ ; and  $\Delta$  is parameter that governs the splitting between levels  $J = 0$  and  $J = 1$ .



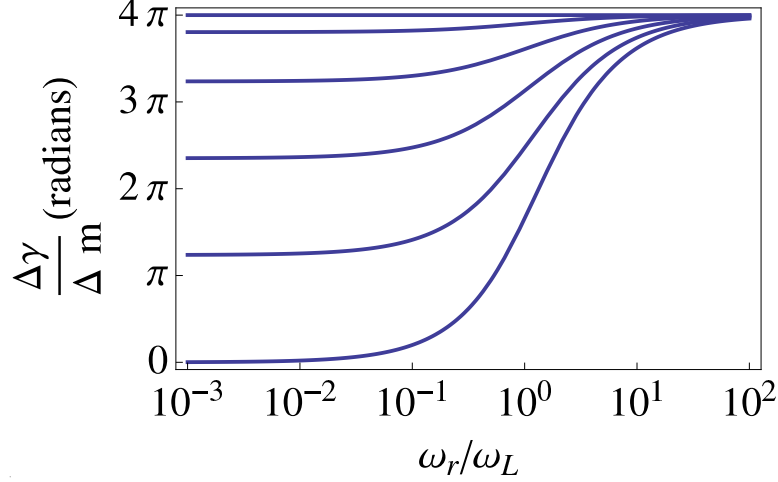


FIG. 2: The extra phase accumulated due to the rotation of the field. In the limit of very fast rotation,  $\omega_r \gg \omega_L$ , the system accumulates a phase of  $4\pi$ , which is unobservable. In this limit, the states are best represented by projections onto the axis of rotation. The various lines represent values of  $\theta_r$  between  $\pi/2$  (bottom line) and 0 (top line) in steps of  $\pi/10$ . As can be seen, when  $\theta_r$  is zero, there is no measurable phase shift, since there is no enclosed solid angle.

The Hamiltonian (57) can be represented by a  $4 \times 4$  matrix, in the basis  $\{|(j_1 j_2) J M_J\rangle\} = \{ |(\frac{1}{2} \frac{1}{2}) 0 0\rangle, |(\frac{1}{2} \frac{1}{2}) 1 1\rangle, |(\frac{1}{2} \frac{1}{2}) 1 0\rangle, |(\frac{1}{2} \frac{1}{2}) 1 -1\rangle \}$

$$H_{\text{dressed}} = \begin{pmatrix} -\frac{3\Delta}{4} & 0 & \frac{1}{2}(\omega_1 - \omega_2) & 0 \\ 0 & \frac{\Delta}{4} + \omega_Z - \omega_r \cos(\theta_r) & \frac{\omega_r}{\sqrt{2}} \sin(\theta_r) & 0 \\ \frac{1}{2}(\omega_1 - \omega_2) & \frac{\omega_r}{\sqrt{2}} \sin(\theta_r) & \frac{\Delta}{4} & \frac{\omega_r}{\sqrt{2}} \sin(\theta_r) \\ 0 & 0 & \frac{\omega_r}{\sqrt{2}} \sin(\theta_r) & \frac{\Delta}{4} - \omega_Z + \omega_r \cos(\theta_r) \end{pmatrix}, \quad (58)$$

where  $\omega_Z = \frac{1}{2}(\omega_1 + \omega_2)$ , is the average of the individual Larmor frequencies. The first item to note is that if the two spins have identical Larmor frequencies,  $\omega_1 = \omega_2$ , then this Hamiltonian is equivalent to that of a spin-0 particle and a spin-1 particle that are independent of each other, there is no coupling between the two states. Each would then evolve according to the previous section on pure spins. This would be the case for the singlet and triplet excited states of the helium atom,  $(1s2s)^{1,3}S$  state, for example. However, should these spins be different from one another (such that  $\omega_1 \neq \omega_2$ ) then coupling corrections arise.

The ordinary adiabaticity criterion specifies that the rotational frequency  $\omega_r$  be small as compared to the Larmor precession frequency  $\omega_L$ , which in this example is given by

$\omega_Z$ . However, now it becomes also necessary to specify whether the Larmor frequency itself is large or small compared to the splitting  $\Delta$  between adjacent  $J$ -levels. This is because the Berry phase arises from a correction to the eigenvalues of the dressed Hamiltonian relative to the non-rotating Hamiltonian. It is therefore worthwhile to cast the non-rotating Hamiltonian in the basis in which it is as diagonal as possible. In the following sections we treat the two limits separately. Since our emphasis here is on the Berry-phase limit, we consider only the limit where  $\omega_r \ll \omega_Z$ , where the rotation rate of the field is small compared to the Larmor frequency. The resulting phase shifts are of course implicit in the theory, however.

#### A. Weak magnetic field, $\omega_r \ll \omega_Z \ll \Delta$

In the low-field limit, but assuming that each Larmor frequency  $\omega_i$  is still far larger than the rotational frequency  $\omega_r$ , we can write down expressions for the energy quite simply. Note that in the absence of rotation, the leading-order energy shift is the sum of the Larmor frequencies themselves, i.e.,  $(1/2)(\omega_1 + \omega_2)M_J = \omega_Z M_J$ . Leaving this correction on the diagonal to break the degeneracy of the  $J = 1$  level, we now treat as perturbations the difference  $(1/2)(\omega_1 - \omega_2)$  and the rotation rate  $\omega_r$ .

Doing so, the leading-order correction due to rotation of the field is given by the diagonal terms in (58) that contain the rotation rate  $\omega_r$ . This correction is the usual Berry phase found above,

$$\gamma^{(0)}(|J M_J\rangle) = -2\pi M_J \cos \theta_r, \quad (59)$$

and it depends on the atomic state only through the total projection of angular momentum  $M_J$ . Thus the ordinary Berry phase in the limit of zero rotation rate is still intact, and is independent of the internal structure.

However, the higher order corrections do depend on this structure. To leading order in the rotation frequency  $\omega_r$ , we find a correction to the Berry phase in the  $|(\frac{1}{2} \frac{1}{2}) 1 1\rangle$  state:

$$\gamma^{(1)}(|11\rangle) = 2\pi \frac{\omega_r}{2\omega_Z} \sin^2(\theta_r) \left( 1 + \frac{(\omega_1 - \omega_2)^2}{4\omega_Z(\Delta + \omega_Z)} \right). \quad (60)$$

The first term is the usual first order correction for a structureless particle (cf. (49)), with the replacement of  $\omega_L$  by  $\omega_Z$ . This should be expected since the energy splitting between

the two states is given by  $\omega_Z$ , and thus is what must be overcome by the rotating field that couples together the differing projections. The second term in the parentheses depends on how strongly the rotating field couples states with differing total  $J$ , as manifested by  $\omega_1 - \omega_2$ . This new correction arises from 4<sup>th</sup>-order mixing in perturbation theory, it is nevertheless linear in the adiabaticity parameter  $\omega_r/\omega_Z$ . A similar expression is found for the  $|1 - 1\rangle$  state,

$$\gamma^{(1)}(|1 - 1\rangle) = -2\pi \frac{\omega_r}{2\omega_Z} \sin^2(\theta_r) \left( 1 - \frac{(\omega_1 - \omega_2)^2}{4\omega_Z(\Delta - \omega_Z)} \right). \quad (61)$$

As is expected from the pure spin case, this state picks up an overall negative sign. However, due to slight changes introduced by the structure, we find a slightly different correction to the second term in parentheses. In fact, we can write down an expression that encapsulates the first order (in  $\omega_r/\omega_Z$ ) correction as

$$\gamma^{(1)}(|J M_J\rangle) = 2\pi M_J \frac{\omega_r}{2\omega_Z} \sin^2(\theta_r) \left( 1 + M_J \frac{(\omega_1 - \omega_2)^2}{4\omega_Z(\Delta + M_J\omega_Z)} \right). \quad (62)$$

While the first term is exactly of the form in (49), the second term describes how the distant  $|J = 0, M_J = 0\rangle$  state affects the accumulated first order phase  $\gamma_{M_J}^{(1)}$ ; namely that the quadratic Zeeman shift in the two  $M_J = 0$  levels distorts the system such that the  $|J = 1, M_J = 1(-1)\rangle$  state is affected more (less) by the  $|J = 1, M_J = 0\rangle$  state.

The two states with  $M_J = 0$  do not acquire a geometric phase at lowest order in  $\omega_r$ , which is appropriate. In this case the leading order perturbation to the dressed Hamiltonian is  $E_Z^{(\pm)} = \pm[(1/2)(\omega_1 - \omega_2)]^2/\Delta$ , which denotes the quadratic Zeeman shift already present in the non-rotating system, and which does not contribute to the Berry phase  $\gamma$ . The quadratic shift after a period  $\tau = 2\pi/\omega_r$  is the dynamical phase the  $M_J = 0$  would nominally acquire. To the first order in  $\omega_r$  in which there is a correction to the  $|J = 0, M_J = 0\rangle$  state arises in 4<sup>th</sup>-order perturbation theory. It is given by

$$\gamma^{(1)}(|0 0\rangle) = 2\pi \frac{\omega_r}{2\Delta} \sin^2(\theta_r) \frac{2E_Z^{(-)}}{\Delta \left( 1 - \left( \frac{\omega_Z}{\Delta} \right)^2 \right)}, \quad (63)$$

where as always the superscript “1” denotes a correction linear in  $\omega_r$ . Here we find a term that appears similar to the  $M_J = 1$  states, with the exception that it occurs in an  $M_J = 0$  state. We have introduced a new energy scale into the problem by adding  $\Delta$  and this allows the  $|0 0\rangle$  state to acquire a first order Berry phase. However, the strength of this phase is reduced by a term proportional to the ratio of the quadratic Zeeman shift in the lower level

to the spin-spin energy splitting. Given our assumptions, this term — while linear in  $\omega_r$  — is a product of multiple small parameters, and is generally smaller than  $\gamma^{(0)}$  for  $M_J = \pm 1$  states.

For the case of the  $|J = 1, M_J = 0\rangle$  state, there is also a 4<sup>th</sup>-order correction, but it takes a very different form. We find, after much algebra,

$$\gamma^{(1)}(|10\rangle) = -2\pi \frac{\omega_r}{2\omega_Z} \sin^2(\theta_r) \frac{2E_Z^{(+)}}{\omega_Z}. \quad (64)$$

This is very different from the  $|00\rangle$  state correction in (63). The important energy scale is the linear Zeeman shift  $\omega_Z$ . Eqs. (63) and (64) carry an important insight; the energy scale responsible for higher-order Berry phases is different for the two  $M_J = 0$  states. The dominant scale in the  $|00\rangle$  is the spin-spin splitting  $\Delta$ . In the  $|10\rangle$ , the dominant energy scale is the linear Zeeman shift. To first order in  $\omega_r$ , there is a correction to the  $M_J = 0$  states that, while similar to the shift in the  $|M_J| = 1$  states, is reduced in magnitude. This reduction is due to the structure, the structure that provides a quadratic Zeeman shift in the  $M_J = 0$  states. For the lower (upper) level, the correction depends on the relative strength of the quadratic Zeeman shift to the spin-spin splitting (linear Zeeman shift). In the regime considered, both of these contributions are very small. The same ideas apply to the  $F = 0$  and  $F = 1$  hyperfine states of Hydrogen, where the magnetic field is coupling states of the same parity. Briefly,  $\gamma^{(1)}$  is influenced by “nearby”  $M_J = \pm 1$  states for the  $|10\rangle$  level, and comparatively less influenced by the “far away”  $M_J = \pm 1$  levels in the  $|00\rangle$  state.

It is instructive to examine these results for different cases of individual Larmor frequencies. In the case where both particles experience the same Larmor frequency in a field,  $\omega_1 = \omega_2$ , then these first-order corrections reduce to the usual first-order corrections for a structureless spin-1 particle, as in Eqn. (49), and the additional  $M_J = 0$  pieces are zero as well. In another limit where one Larmor frequency dominates the other, say  $\omega_1 \gg \omega_2$ , then  $\gamma^{(1)}$  for the  $|M_J| = 1$  states reduce to the first order correction of the dominant spin alone, reflecting the fact that the weaker spin is coupled to the stronger one and gets dragged along for the ride. This happens, for example, in the  $F = 1$  hyperfine ground state of the hydrogen atom, where the nuclear g-factor is far smaller than the electron g-factor.

### B. Strong magnetic field, $\omega_1, \omega_2 \gg \Delta \gg \omega_r$

In the other limit, where the magnetic field is large compared to the splitting between adjacent  $J$ -levels, it is more useful to construct the dressed Hamiltonian in an alternative basis. Namely, the non-rotating Hamiltonian is more nearly diagonal in the independent-spin basis  $|j_1 m_1 j_2 m_2\rangle$ , where the four Zeeman energies  $E_{m_1, m_2}$  are given simply by  $m_1 \omega_1 + m_2 \omega_2$ :

$$\begin{aligned} E_{\frac{1}{2}, \frac{1}{2}} &= \frac{1}{2}(\omega_1 + \omega_2) \\ E_{\frac{1}{2}, -\frac{1}{2}} &= \frac{1}{2}(\omega_1 - \omega_2) \\ E_{-\frac{1}{2}, -\frac{1}{2}} &= -\frac{1}{2}(\omega_1 + \omega_2) \\ E_{-\frac{1}{2}, \frac{1}{2}} &= -\frac{1}{2}(\omega_1 - \omega_2), \end{aligned} \tag{65}$$

as appropriate to this Paschen-Back limit of the Zeeman effect. This is an example of the aside in Sec. II where we make the *ansatz*  $(\alpha, \beta) e^{-i m_1 \omega_r t} e^{-i m_2 \omega_r t}$ . The remaining Hamiltonian, which includes the rotation of the field and the spin-spin interaction, is recast as follows

$$H_{\text{dressed}} = \begin{pmatrix} E_{\frac{1}{2}, \frac{1}{2}} + \frac{\Delta}{4} - \omega_r \cos(\theta_r) & \frac{\omega_r}{2} \sin(\theta_r) & 0 & -\frac{\omega_r}{2} \sin(\theta_r) \\ \frac{\omega_r}{2} \sin(\theta_r) & E_{\frac{1}{2}, -\frac{1}{2}} - \frac{\Delta}{4} & \frac{\omega_r}{2} \sin(\theta_r) & -\frac{\Delta}{2} \\ 0 & \frac{\omega_r}{2} \sin(\theta_r) & E_{-\frac{1}{2}, -\frac{1}{2}} + \frac{\Delta}{4} + \omega_r \cos(\theta_r) & -\frac{\omega_r}{2} \sin(\theta_r) \\ -\frac{\omega_r}{2} \sin(\theta_r) & -\frac{\Delta}{2} & -\frac{\omega_r}{2} \sin(\theta_r) & E_{-\frac{1}{2}, \frac{1}{2}} - \frac{\Delta}{4} \end{pmatrix} \tag{66}$$

Once again, we can immediately read the Berry-phase contribution from the diagonal components, as

$$\begin{aligned} \gamma^{(0)}(|j_1 m_1 j_2 m_2\rangle) &= -2\pi(m_1 + m_2) \cos(\theta_r) \\ &= -2\pi M_J \cos(\theta_r). \end{aligned} \tag{67}$$

The phase accumulates due to the individual spins separately, as expected when the spins interact weakly with each other compared to their interaction with the field.

This independent accumulation of phase leads to a different interpretation of the  $M_J = 0$  states: in the limit of small magnetic field compared to the spin-spin energy splitting, we had attributed this to an  $M_J = 0$  projection while here, we can attribute this to  $m_1 = \pm \frac{1}{2}$  accumulating  $\pm(\omega_r/2) \cos(\theta_r)$  extra energy and the  $m_2 = \mp \frac{1}{2}$  accumulating  $\mp(\omega_r/2) \cos(\theta_r)$ .

To cement this idea even further, we find there are two independent first-order contributions to the first-order non-adiabatic correction  $\gamma^{(1)}$ , computed first neglecting  $\Delta$ :

$$\gamma^{(1)}(|j_1 m_1 j_2 m_2\rangle) = 2\pi m_1 \frac{\omega_r}{2\omega_1} \sin^2(\theta_r) + 2\pi m_2 \frac{\omega_r}{2\omega_2} \sin^2(\theta_r) \quad (68)$$

$$= \gamma_1^{(1)}(|j_1 m_1\rangle) + \gamma_2^{(1)}(|j_2 m_2\rangle). \quad (69)$$

This is exactly the contribution one would expect from two *independent* spins following a rotating field. When the spins are anti-aligned, or  $m_1 = -m_2$ , we find no correction at this order. It is worth noting that this perturbative expansion breaks down if  $\omega_r \sim \omega_i$ . Thus, should the rotation rate be fast with respect to one of the Larmor frequencies, but not the other, then the measured phase difference cannot be treated perturbatively in this regime.

The explicit effect of internal structure, manifested in the splitting  $\Delta$ , appears as a next-order correction:

$$\gamma_1^{(1)} \rightarrow 2\pi m_1 \frac{\omega_r}{2\omega_1} \sin^2(\theta_r) \left( 1 + \left( \frac{\Delta}{2\omega_2} \right)^2 \right) \quad (70)$$

$$\gamma_2^{(1)} \rightarrow 2\pi m_2 \frac{\omega_r}{2\omega_2} \sin^2(\theta_r) \left( 1 + \left( \frac{\Delta}{2\omega_1} \right)^2 \right). \quad (71)$$

Of course, there are many routes by which 4<sup>th</sup>-order perturbation theory can affect this state. We only give one route which produces a phase shift proportional to  $\omega_r$  after one period of oscillation. There is a structure correction for each non-adiabatic spin that depends on the relative strength of the spin-spin splitting to the other Larmor frequency. Thus, we require that the spin-spin splitting be small compared to each of the Larmor frequencies in order to make this expansion. Again, this is a more restrictive condition on adiabaticity than is usually employed for two independent spins.

## V. POLAR MOLECULES IN A ROTATING ELECTRIC FIELD

Molecules bring yet another degree of freedom to the picture, namely end-over-end rotation with eigenstates  $|N M_N\rangle$ . In addition, if the molecule is polar, it has an electric dipole moment that can be acted upon by a rotating electric field. In this section we will consider only diatomic molecules, and only one of a fairly simple structure, to illustrate how our formalism applies to them. The lowest-order Berry phase was worked out recently in this system [22], but the higher-order corrections are implicit there as well.

For the sake of illustration we choose the simplest of diatomic molecules, a  $^1\Sigma$  molecule with no hyperfine structure. In a rotating electric field this system is described by a Hamiltonian of the form

$$H_{\text{dressed}} = B\vec{N}^2 - \vec{\mu}_m \cdot \vec{\mathcal{E}} - \omega_r(\cos(\theta_r)N_z - \sin(\theta_r)N_x), \quad (72)$$

where  $\vec{N}$  is the end-over-end rotational angular momentum of the molecule,  $\mu_m$  is the electric dipole moment of the molecule and  $\mathcal{E}$  is the electric field strength. Since we are in the frame of the electric field and the electric dipole moment points along the molecular axis, we do not have any couplings of  $M_N$  or  $\Lambda$ , where  $\Lambda$  is the projection of total angular momentum onto the internuclear axis. For  $\Sigma$ -molecules, this means there are no couplings to excited electronic states by the applied electric field at the low fields we consider.

For simplicity, we consider here the coupling only between the  $N = 0$  and  $N = 1$  rotational levels of the molecule, assuming weak coupling of rotational states due to the electric field, i.e.,  $\mu_m\mathcal{E} \ll B$ . The formalism can of course be extended to arbitrarily large  $N$  values as needed. It is nice to note that this formalism has an atomic analog: the  $^1S_0$  and  $^1P_1$  states of noble gas and alkaline-earth atoms have opposite-parity and are coupled by the Stark interaction. This approach gives the corrections for states of opposite parity coupled by the Stark interaction. The dressed Hamiltonian reads, in the basis  $\{|NM_N\rangle\} = \{|00\rangle, |1-1\rangle, |10\rangle, |1+1\rangle\}$

$$H_{\text{dressed}} = \begin{pmatrix} 0 & 0 & -\frac{1}{\sqrt{3}}\mu_m\mathcal{E} & 0 \\ 0 & 2B - \omega_r \cos(\theta_r) & \frac{1}{\sqrt{2}}\omega_r \sin(\theta_r) & 0 \\ -\frac{1}{\sqrt{3}}\mu_m\mathcal{E} & \frac{1}{\sqrt{2}}\omega_r \sin(\theta_r) & 2B & \frac{1}{\sqrt{2}}\omega_r \sin(\theta_r) \\ 0 & 0 & \frac{1}{\sqrt{2}}\omega_r \sin(\theta_r) & 2B + \omega_r \cos(\theta_r) \end{pmatrix} \quad (73)$$

Note the the electric Hamiltonian is off-diagonal in the basis of parity eigen-states. In the absence of the perturbation  $\omega_r$ , this Hamiltonian appears to have a complete degeneracy among the three states with  $N = 1$ . In the magnetic field case above, this degeneracy was broken by the linear Zeeman effect acting on the diagonal matrix elements. To achieve the same feat here, we must account for the off-diagonal mixing due to electric field. Note the similarity of this procedure to that of Vutha and DeMille [22].

We first diagonalize the  $M_N = 0$  subspace, using the mixing angle  $\delta$  defined by

$$\tan(\delta) = -\frac{\mu_m\mathcal{E}}{\sqrt{3}B} = -x, \quad (74)$$

with the usual eigenvectors  $(\cos(\delta/2), \sin(\delta/2))$  and  $(-\sin(\delta/2), \cos(\delta/2))$ . The explicit values in terms of the parameter  $x$  are

$$\cos\left(\frac{\delta}{2}\right) = \sqrt{\frac{\sqrt{1+x^2}+1}{2\sqrt{1+x^2}}} \quad (75)$$

$$\sin\left(\frac{\delta}{2}\right) = \sqrt{\frac{\sqrt{1+x^2}-1}{2\sqrt{1+x^2}}}. \quad (76)$$

In terms of this mixing angle the transformed Hamiltonian, with electric-field-dependent terms on the diagonal only, reads

$$H_{\text{dressed}} = \begin{pmatrix} B(1 - \sqrt{1+x^2}) & -\frac{\omega_r}{\sqrt{2}} \sin(\frac{\delta}{2}) \sin(\theta_r) & 0 & -\frac{\omega_r}{\sqrt{2}} \sin(\frac{\delta}{2}) \sin(\theta_r) \\ -\frac{\omega_r}{\sqrt{2}} \sin(\frac{\delta}{2}) \sin(\theta_r) & 2B - \omega_r \cos(\theta_r) & \frac{\omega_r}{\sqrt{2}} \cos(\frac{\delta}{2}) \sin(\theta_r) & 0 \\ 0 & \frac{\omega_r}{\sqrt{2}} \cos(\frac{\delta}{2}) \sin(\theta_r) & B(1 + \sqrt{1+x^2}) & \frac{\omega_r}{\sqrt{2}} \cos(\frac{\delta}{2}) \sin(\theta_r) \\ -\frac{\omega_r}{\sqrt{2}} \sin(\frac{\delta}{2}) \sin(\theta_r) & 0 & \frac{\omega_r}{\sqrt{2}} \cos(\frac{\delta}{2}) \sin(\theta_r) & 2B + \omega_r \cos(\theta_r) \end{pmatrix} \quad (77)$$

In the limit that  $x \ll 1$  we see that the diagonal terms for the two  $M_N = 0$  states are merely the quadratic Stark shift  $E_S^{(\pm)} = \pm(\mu_m \mathcal{E})^2/6B$ . From here we can read off the ordinary Berry phase from the diagonal perturbations linear in  $\omega_r$ , yielding the usual

$$\gamma^{(0)}(|\tilde{N} M_N\rangle) = -2\pi M_N \cos(\theta_r), \quad (78)$$

where by  $\tilde{N}$  is meant the appropriate eigenstate of the field-mixed  $M_N = 0$  states [22]. This diagonalization removed the degeneracy of the  $M_N = 0$  level with the  $M_N = \pm 1$  levels of the  $N = 1$  subspace. However, the degeneracy among the  $M_N = \pm 1$  states still exists.

Having quasi-broken the degeneracy in the  $N = 1$  levels, we can now evaluate the first-order adiabatic correction term using standard second-order perturbation theory. It is evident that both states  $|N, M_N = \pm 1\rangle$  experience the same additional phase at this order (due to their degeneracy in the non-perturbing Hamiltonian), given by

$$\gamma^{(1)}(|1, \pm 1\rangle) = -\pi \left( \frac{6B}{(\mu_m \mathcal{E})^2} \right) \omega_r \sin^2(\theta_r), \quad (79)$$

where an expansion in the small parameter  $x$  has been applied. Requiring this to be a small correction identifies the adiabaticity criterion for this situation. If this case were analogous to the magnetic field case, we would only be concerned about the magnitude of  $\omega_r$  with respect to  $\mu_m \mathcal{E}$ , which is the stand-in for the Larmor frequency in this case. However, Eqn. (79) suggests a slightly different criterion, namely  $\omega_r B \ll (\mu_m \mathcal{E})^2$  must hold in order



to recover the simple leading-order Berry phase. To understand the origin of this criterion we look at the term in the large parentheses in (79). It is the inverse of the Stark energy in the absence of field rotation for the upper level. We can rewrite (79) as

$$\gamma^{(1)}(|N M_N = \pm 1\rangle) = -2\pi \frac{\omega_r}{2 E_S} \sin^2(\theta_r), \quad (80)$$

and we recover a form reminiscent of the pure spin case (cf. (49)), where the Larmor frequency,  $\omega_L$ , is replaced by  $E_S$ , the quadratic Stark shift. In order to be an adiabatic correction, it is immediately evident why  $\omega_r B \ll (\mu_m \mathcal{E})^2$  must hold; the rotation rate must be small compared to the energy splitting in that level. In this case, the splitting is quadratic in electric field and therefore a secondary energy scale — the rotational level splitting or internal structure — must come into play.

By similar reasoning, we can arrive at the  $\gamma^{(1)}$  corrections for the  $M_N = 0$  states as well. That of the lower level is given by

$$\gamma^{(1)}(|\tilde{N} \sim 0, 0\rangle) = -2\pi \frac{\omega_r}{2B} \sin^2(\theta_r) \frac{E_S}{2B}, \quad (81)$$

where we find the requirement that  $\omega_r E_S \ll B^2$  must hold. In this case, our assumptions clearly support this adiabatic criterion since we are in the regime  $\omega_r \ll \mu_m \mathcal{E} \ll B$ . This correction is linear in  $\omega_r$ , but suppressed by the ratio of the Stark energy to the rotational constant of the molecule. Physically, this is because this state is far removed from the “degeneracy” in the  $N = 1$  levels. It is the electric analogue of the weak magnetic field limit of the spin-spin interaction. The correction for the upper level is given by

$$\gamma^{(1)}(|\tilde{N} \sim 1, 0\rangle) = 4\pi \frac{\omega_r}{2 E_S} \sin^2(\theta_r). \quad (82)$$

This correction is in the opposite direction to and twice that of the  $|N M_N = \pm 1\rangle$  states because the  $|\tilde{N} \sim 1, 0\rangle$  state is influenced by the two states,  $M_N = \pm 1$ , that are below it in energy. This is in contrast to the coupled spins in a magnetic field case, where each  $M_N = \pm 1$  contributed equally in magnitude but opposite in sign. This is due to the lack of any linear Stark shift in the  $M_N = \pm 1$  levels.

It is evident that polar molecules in a rotating electric field are quite similar to magnetic dipoles in rotating magnetic fields. There is an energy splitting in comparison with which the rotation of the field must be small to ensure adiabaticity. If there is a shift in energy that is linear with the applied field, then the rotation rate must be small compared to this

energy. However, if the energy scales quadratically with the applied field, the rate of rotation must be small in comparison to the energy shift in the field. Thus, the internal structure is quite important in regimes of quadratic field shifts and introduces different adiabaticity requirements on  $\omega_r$  in terms of the applied field and internal structure.

## VI. CONCLUSIONS

The dressed-state formalism is a natural way to treat quantum mechanical objects subject to time-periodic driving such as we have considered here. It allows for the inclusion of arbitrary internal structure of the object, and still reveals the exact dressed eigen-energies at arbitrary rotation rates. In the limit of slow rotations, it also reduces, as it must, to the usual geometric Berry phase. Because it includes the structure of the atom or molecule considered, however, it is also able to shed light on the influence of this structure on non-adiabatic corrections to the geometric phase. It is therefore expected to be a powerful tool to be used when analyzing high-precision spectroscopic data in the presence of periodic driving [30, 31].

The treatment herein has considered only the simplest case of a magnetic or electric field whose direction precesses uniformly about a given axis. It is to be expected, however, that this treatment is yet more general, and that dressed states for arbitrary periodic driving Hamiltonians could be constructed, at least numerically. It could, for example, be combined with the Floquet analysis that has been used previously for unstructured particles [14, 15, 16, 32].

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## APPENDIX A

In Sec. III, we had a Hamiltonian of the form

$$H = \omega_L S_z - \omega_r (\cos(\theta_r) S_z - \sin(\theta_r) S_x), \quad (\text{A1})$$

which looks like

$$H = \begin{pmatrix} a_1 & c_1 & 0 & 0 & \dots \\ c_1 & a_2 & c_2 & 0 & \vdots \\ 0 & c_2 & \ddots & \ddots & c_{n-2} \\ \vdots & 0 & c_{n-2} & a_{n-1} & c_{n-1} \\ \dots & 0 & 0 & c_{n-1} & a_n \end{pmatrix}, \quad (\text{A2})$$

In our system, not only is this matrix symmetric, it contains the following added symmetry:  $a_1 = -a_n$ ,  $a_2 = -a_{n-1}$ , etc. Also, the coupling coefficients  $c_i$  follow a similar property:  $c_1 = c_{n-1}$ , etc. These properties are key to simplifying the eigenvalues of the tridiagonal matrix in our case.

Eigenvalues of an  $n \times n$  tridiagonal matrix are given by the roots of the polynomial  $p_n$ , defined recursively by

$$\begin{aligned} p_0(\lambda) &= 1, \\ p_1(\lambda) &= (a_1 - \lambda), \\ p_2(\lambda) &= (a_2 - \lambda)p_1(\lambda) - c_1^2 p_0(\lambda) \\ &\vdots \\ p_n(\lambda) &= (a_n - \lambda)p_{n-1}(\lambda) - c_{n-1}^2 p_{n-2}(\lambda). \end{aligned} \quad (\text{A3})$$

In our problem, the constants are defined as

$$a_m = m(\omega_L - \cos(\theta_r)), \quad (\text{A4})$$

$$c_m \sim \begin{pmatrix} S & 1 & S \\ -m & q & m - q \end{pmatrix}. \quad (\text{A5})$$

Thus, the symmetry pops right out.

A simple example is the case of  $S = 1/2$ . Here we find that the characteristic polynomial is

$$p_2(\lambda) = (\lambda - \frac{1}{2}a)(\lambda + \frac{1}{2}a) - \left(\frac{1}{2}b\right)^2 = (\lambda^2 - \frac{1}{4}(a^2 + b^2)) \quad (\text{A6})$$

where  $a = (\omega_L - \omega_r \cos(\theta_r))$  and  $b = \omega_r \sin(\theta_r)$ . For the case of  $S = 1$  we find a similar equation (after simplification)

$$\begin{aligned} p_3(\lambda) &= \lambda((\lambda - a)(\lambda + a) - 2\left(\frac{1}{\sqrt{2}}b\right)^2) \\ &= \lambda(\lambda^2 - (a^2 + b^2)) \end{aligned} \quad (\text{A7})$$

For integer values there is always a diagonal element that is 0. As is evident, this has the same form as (A6) with the added piece of  $\lambda$  multiplying everything yielding an eigenvalue of 0. In addition, Eq. (A7) is scaled by a factor of 4 from from Eq. (A6), thus making the the eigenvalues a factor of 2 larger. This is because the value of  $m$  in (A7) is twice as large as the value of  $m$  in (A6).

We can generalize the characteristic polynomial to a very simple expression due to the added symmetries. It is given by

$$p_{2S+1} = \prod_{m=(m_{\min} \geq 0)}^{m_{\max}} (\lambda^{2(1-\delta_{m_{\min},0})} - m^2(a^2 + b^2)). \quad (\text{A8})$$

The Kronecker  $\delta$ -function in (A8) is to insure that in the event  $m_{\min} = 0$  there is only one eigenvalue  $\lambda = 0$ .

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